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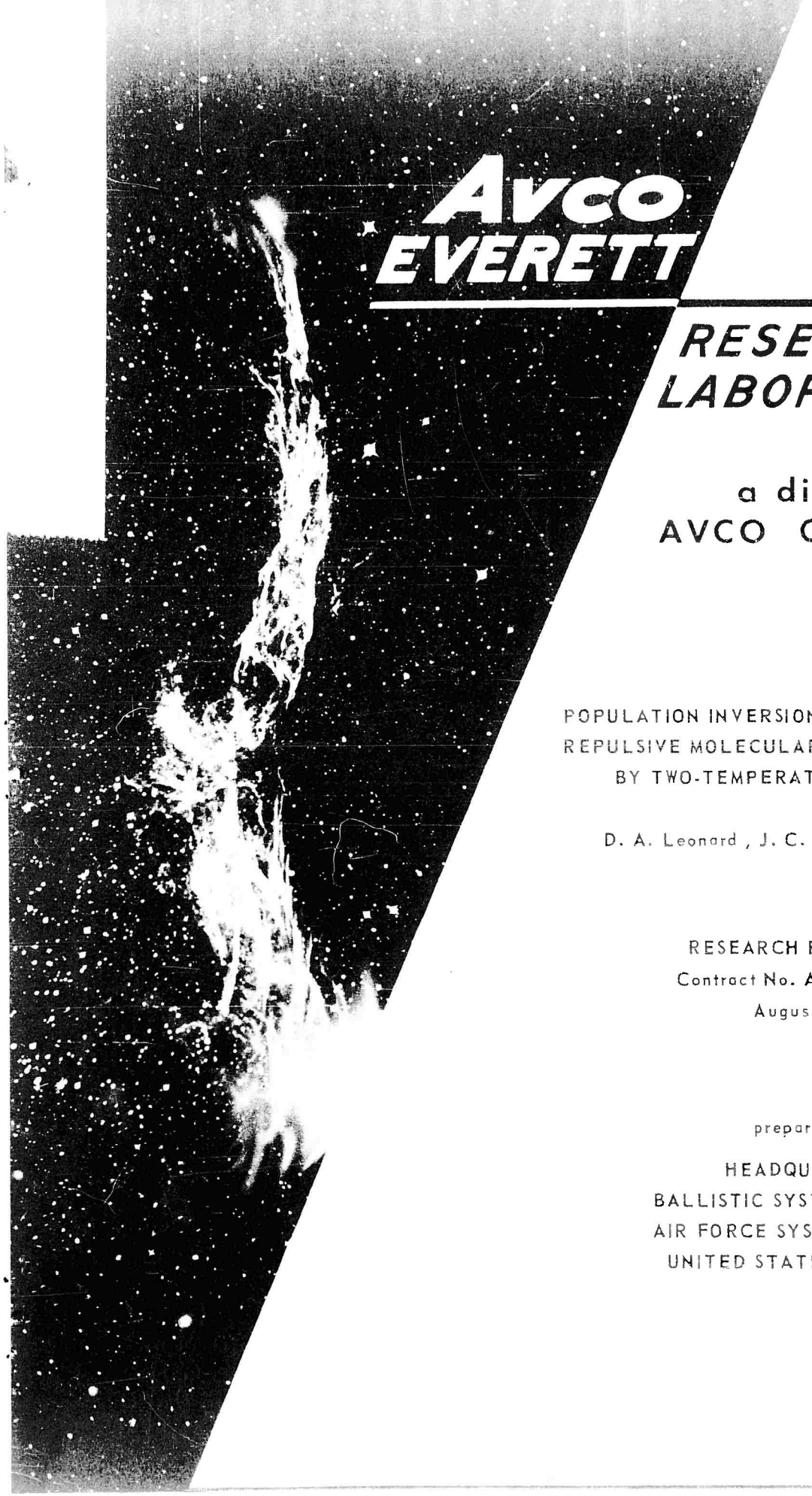

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POPULATION INVERSION BETWEEN BOUND AND REPULSIVE MOLECULAR ELECTRONIC STATES BY TWO-TEMPERATURE EQUILIBRIUM

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ABSTRACT

The well known ultraviolet emission continuum of molecular hydrogen which results from transitions between the $a^3\Sigma_g^+$ and $b^3\Sigma_u^+$ electronic states is suggested as a transition for a high power, ultraviolet, gas laser. In an electric arc, collisions of hot electrons with ground state molecules populate these two states. The populations can be inverted because of the favorable ratio of lifetimes of the two states. The upper state has a radiative lifetime of about 10^{-8} sec; the lower state is repulsive and dissociates in less than 10^{-13} sec. Steady state inversion can be achieved by maintaining the heavy particle temperature below the electron temperature and by maintaining the hydrogen atom concentration below the equilibrium value corresponding to the electron temperature. This suppresses the population of the lower state by reducing the rate of recombination of ground state atoms into the repulsive state. A two temperature quasi-equilibrium model requires a temperature ratio of 1.5 for inversion. In preliminary experiments with a hydrogen arc running at 5 mm Hg pressure, steady-state gas conditions were achieved from which $a^3\Sigma_g^+ - b^3\Sigma_u^+$ spontaneous emission at 3500 Å of 40 watts/cm^3-μ-ster was measured and a gain of .2% per meter and an inversion ratio of 10^4 were calculated.

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This letter proposes a new class of gas laser systems, well suited for high power operation, in which the upper energy level of the laser transition is predominately populated by direct electron impact at the electron temperature, and the lower energy level of the laser transition is controlled by collisions between gas atoms at the heavy particle temperature. An example of such a system for direct two-temperature operation is a molecule that has an upper bound electronic state and a lower repulsive electronic state, for example the bound $a^3\Sigma_g^+$ and the repulsive $b^3\Sigma_u^+$ states of hydrogen, which are shown in Fig. 1. The criterion for inversion is that the electron temperature be made sufficiently greater than the gas temperature, a condition found in high current density arcs running under steady-state conditions at moderate pressure.

For the case of hydrogen, radiation from "a" to the ground state is not allowed. The "a" to "b" transition is the only allowed radiative transition from the "a" state.[1] [2] Population inversion between "a" and "b" is achieved because of the very short lifetime of the "b" state which dissociates spontaneously. The lifetime of the "b" state can be approximately calculated from the acceleration the hydrogen atoms receive from the repulsive potential. This time can be expressed as $\tau\,(b) = (2MV_b)^{1/2}\,(dV/dr)^{-1}$, where V_b is the energy measured up from the dissociation

limit of the "X" state and dV/dr is evaluated at V_b. For example, at the internuclear separation corresponding to the minimum of the "a" state, $\tau(b)$ has the value 10^{-14} sec.

A simple two-temperature calculation has been made in order to estimate the inversion which could be achieved in the hydrogen system under certain circumstances. In this model, the following processes are assumed in equilibrium:

$$H_2 \quad (a^3\Sigma_g) \quad \overset{T_e}{\rightleftharpoons} \quad H_2 \quad (X^1\Sigma_g)$$

$$H_2 \quad (b^3\Sigma_u) \quad \overset{T}{\rightleftharpoons} \quad 2\,H \quad (1s)$$

$$H_2 \quad (X^1\Sigma_g) \quad \overset{T}{\rightleftharpoons} \quad 2\,H \quad (1s)$$

where T_e is the electron temperature, and T is the gas temperature. If these equilibria are satisfied, the ratio of "a" to "b" state populations is given as $[H_2(a)]/[H_2(b)] = \exp(E_b/T - E_a/T_e)$. $[H_2(b)]$ is defined as the average concentration of molecules in the repulsive state at the same internuclear separation as the $a^3\Sigma_g$ for a particular frequency of radiation. The criterion for inversion is then that $T_e/T > E_a/E_b = 1.50$ for the v = 0 level, and $E_a/E_b = 1.34$ and 1.27 for the small internuclear separation side of the "a" state potential curve for the v = 1 and v = 2 levels, respectively. Figure 2 is a plot of $[H_2(a)]/[H_2(b)]$ vs T_e/T at various values of T_e for the v = 0 level of the "a" state, and indicates that enormous inversions are possible for relatively modest values of T_e/T.

The radiation from this "a" to "b" transition appears as a broad continuum extending from 1800 Å to 5000 Å. Assuming a large inversion ratio, the fractional gain per unit length $\Delta I/I\Delta x$ can be expressed[3] as $\Delta I/I\Delta x = \lambda^5 P(\lambda)/4\pi \hbar c^2$ where $P(\lambda)$ is the spontaneous radiation power per unit volume, wavelength, and solid angle. In preliminary arc experiments at 5 mm Hg pressure of H_2 and 500 amps/cm^2, we have

observed $P(\lambda = 3500\ \text{Å}) = 40$ watts/cm^3-μ-ster, which would correspond to a gain of about 0.2% per meter. We also estimate theoretically from the rate equations and from radiation measurements that under the conditions of the experiment $T_e = 3$ ev and $T_e/T = 7$. This implies the large inversion ratio of 9×10^4, limited by direct electron excitation from "X" to "b". We expect practical gains of a few percent to be achievable at higher pressure and current density, at the expense of less population inversion. The threshold power for laser oscillation of a system of this kind is at a power level in the kilowatt range. Correspondingly, very high saturation power should also be possible.

Many other bound-repulsive molecular systems exist,[1] such as He_2, Ne_2, Ar_2, Kr_2, Xe_2, Hg_2 and Cd_2. The first five radiate in the vacuum UV and all seven have repulsive ground states, unlike hydrogen. Many molecules may also have favorable bound-repulsive state pairs, for which the repulsive state does not dissociate into two ground state atoms. Hydrogen was chosen for our preliminary investigation because its properties are better known and it is experimentally easiest to handle.

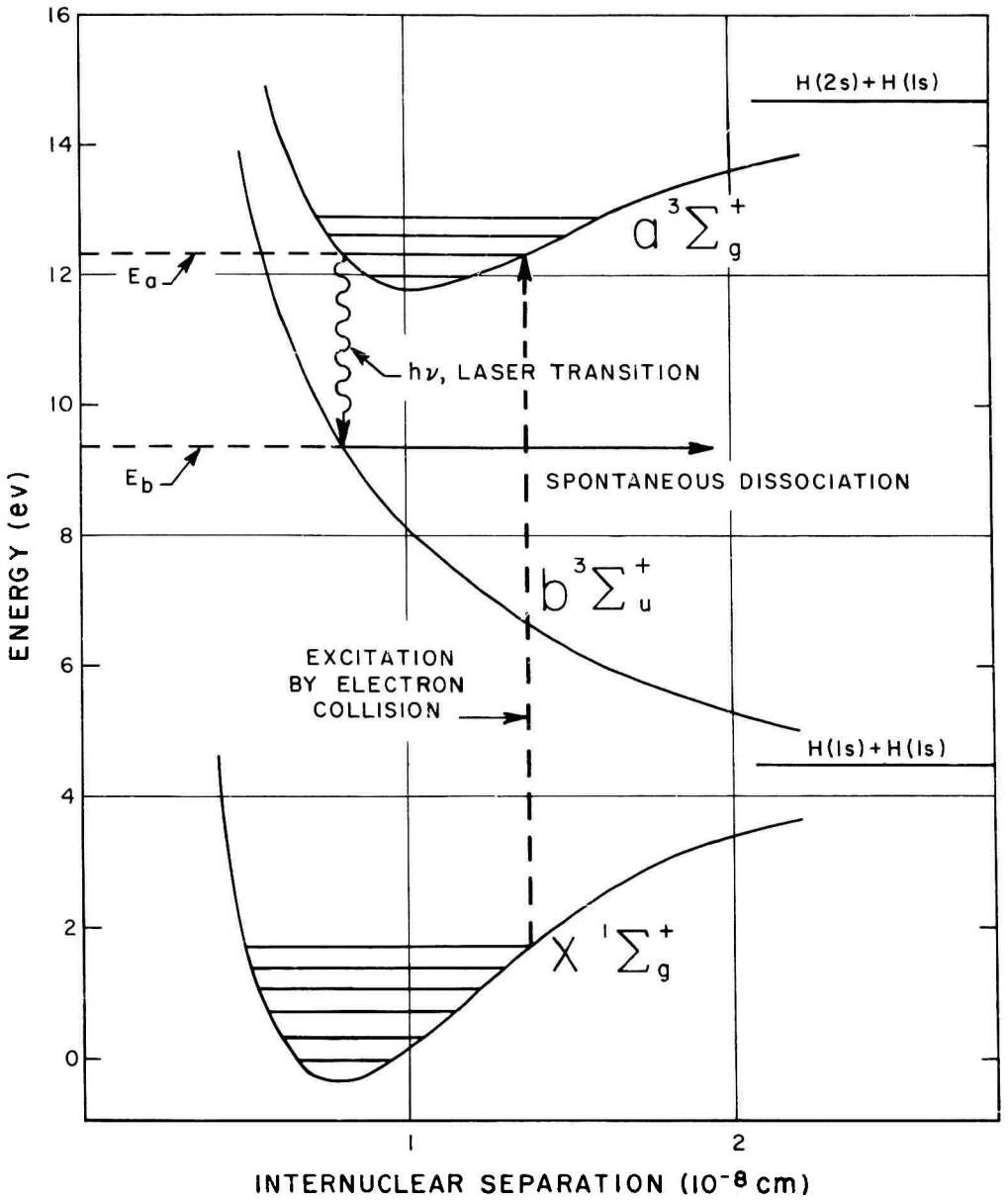

Fig. 1 The potential energy diagram for molecular hydrogen showing the ground state "X" and the "a" and "b" triplet electronic states, which give rise to the continuous spectrum of hydrogen, long used as a light source in UV absorption spectroscopy. The radiative lifetime of the "a" state has been theoretically calculated as 10^{-8} sec.(2) The "b" state spontaneously dissociates in about 10^{-14} sec. It is, therefore, possible to invert the "a" and "b" populations even when collision times with electrons become much shorter than the radiative lifetime of the upper state. A typical excitation, radiation, and dissociation path is shown.

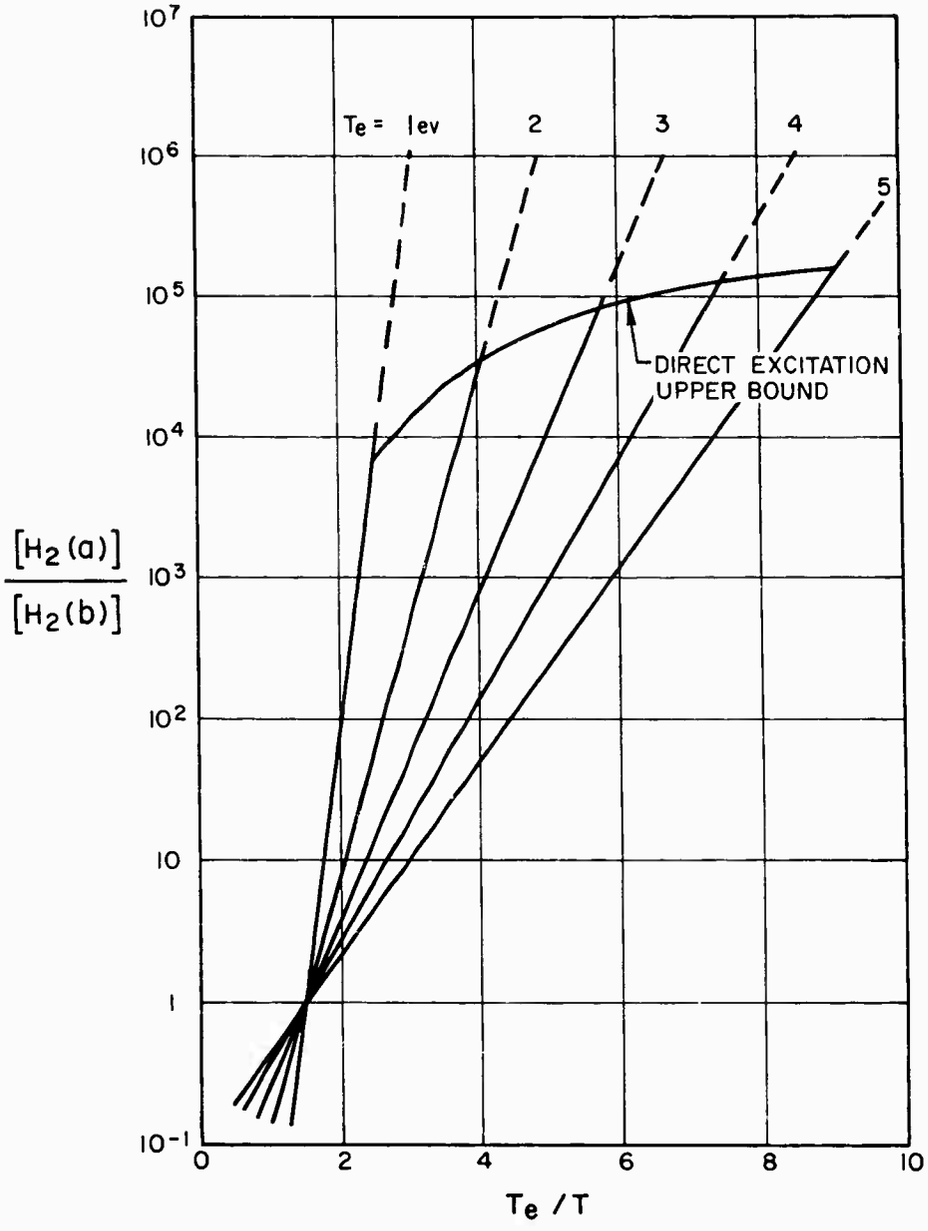

Fig. 2 The population inversion ratio between the "a" and "b" states as a function of the ratio of the electron temperature to the gas temperature at various electron temperatures. The curved line is an estimate from published cross-sections[4] of the upper bound to the inversion ratio because of direct excitation from "X" to "b" by electron impact.

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